SAFETY ANALYSIS OF AN IRRADIATION DEVICE FOR ⁹⁹Mo PRODUCTION IN RA-3 REACTOR

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ABSTRACT

The Argentine RA-3 research reactor (5 MW) has been converted to LEU fuel more than nine years ago. Since then, it has been operating with LEU fuel, which has been designed and fabricated at the National Atomic Energy Commission (CNEA). The Nuclear Regulatory Authority (ARN) is the institution in charge of the installation safety control. It is under this framework that the ARN has elaborated a neutronic calculation model for the RA-3 core, paying special attention to the device presently used for the irradiation of (HEU) ²³⁵U targets required to obtain ⁹⁹Mo as a fission product. A regulatory analysis of results is carried out in the framework of ARN standards for fixed experiments. For such purpose, calculated reactivity values associated with such device are compared with recently measured values at the installation. Finally, and according to guidelines established in the first part of this work, a calculation model for a new device proposed by CNEA for the irradiation of metallic (LEU) uranium targets and still at its design stage, is here analysed.

1 INTRODUCTION

The RA-3 is a MTR reactor with plate type standard fuel elements (SFE). It is located at the surroundings of Buenos Aires city, Argentina, and it operates at a maximum of 5 MW, being radioisotope production its main objective. It is fuelled with U_3O_8 -Al (LEU) fuel since 1990. The SFE contains 290.7 g 235 U distributed in 19 plates, while its control fuel element (CFE) has only 14 plates (identical to those of the SFE), i.e. it contains 214.2 g 235 U.

Although the core configuration may be modified, it usually remains fixed with 25 FE, being 21 of them SFE and the remaining 4 of the CFE type. It operates having an equilibrium burnup distribution, reloading 1 FE per cycle of 20 fpd approximately, with a discharge burnup of about 45% of 235 U consumed. A central irradiation channel is designed to provide a maximum (nonperturbed) thermal flux of 1.6×10^{14} n/cm² s approximately, in order to irradiate 235 U targets to produce 99 Mo as a fission product.

CNEA is both the owner and operator of the RA-3 reactor. Since 1995 the ARN is the independent organisation in charge of radiological and nuclear safety control of nuclear installations, and it is under the framework of the ARN that this work has been carried out.

The purpose of this work is to elaborate a calculation system for the RA-3 reactor, independent from the operator's system, capable of calculating all the interesting situations as well as verifying compliance with operational limits and conditions each time any non-routine modifications are proposed, such as:

- Irradiation of prototype U₃Si₂-Al FE having non-standard ²³⁵U loading.
- Replacement of the present HEU targets used for ⁹⁹Mo production with two possible LEU targets, one of them of UAl₂-Al and the other constituted by metallic U foils.

Emphasis is here put in the description of the core perturbation produced by the introduction of such targets, and the device modelling is thus described in detail. The rest of the core model is briefly shown. Finally, calculated values are compared with measurements carried out by CNEA /1/, showing compliance with ARN standards of application.

2 NEUTRONIC CALCULATION MODELS

2.1 Core model

The core model is here briefly presented, excluding the irradiation device (ID), explicitly described in next section.

Two calculation codes were used: the well-known WIMS lattice code /2/ used to generate cross section (XS) and CITATION /3/ in its improved version CITVAP /4/, both of them run in the environment of MTR_PC System /5/.

The core was modelled in a 3-D geometry and a 2-group energy structure was used for the reactor calculation. Cell calculations were performed with WIMS with its usual full-library energy structure (69 groups) and a "fewgroup" structure of 14 groups, using a 1-D geometry.

Two different reactor states were used as reference: the here called C93_EOC and C94_BOC, corresponding to end-of-cycle and beginning-of-cycle cores number 93 and 94 respectively. Every core configuration is described using the same geometric model, being burnup distribution and control rod (CR) insertion their only difference. Between configurations 93 and 94 the reactor experienced a long shutdown, so that most of the low power measurements were carried out during that period for the C94_BOC in cold conditions.

Figure 1 shows a horizontal core cross section at half height. Each grid position is represented by a rectangle of 7.7 cm x 8.1 cm. Each FE is modelled having three materials, two of them the frames and the central one the "meat". Along the axial dimension fuel channels are divided into 10 regions, each of them having an associated burnup. The CFE has been modelled as the SFE except that two additional channels for the absorbing plates have been included. In this case, the 10-region partition enables partial insertion of absorbers. Irradiation boxes, graphite reflector and water reflector have been modelled as homogeneous non-subdivided regions located in a grid position. The central irradiation box is described separately.

For the sake of shortness no details will be given of WIMS calculations for XS generation. It will only be pointed out that three basically different models have been used:

- Plate SFE model (5 slabs) from which XS for either fuel region or frames are obtained.
- Plate model (12 slabs) for the channel having absorbing plates surrounded by fuel, in two different states; ABSORBER-IN or OUT.
- Cylindrical model (9 annuli) in which the whole reactor is represented by: a central irradiation box, homogenised fuel and moderator, graphite reflector, water reflector and some intermediate structural zones. XS for the central empty (water) region, as well as graphite and water reflector XS for an average fuel burnup are obtained from this model.

2.2 ⁹⁹Mo irradiation device model

For the sake of safety and simplicity in handling, the ID was designed in such a way that when it is withdrawn, the central grid position is not accessible for a FE. An empty box (EB) is located inside this position, having the same outer dimensions as a SFE and a 3mm thick wall. At its lower end a bottom nozzle similar to that of the FE fixes it to the reactor grid. The EB always remains inserted, whether the reactor is operating or not, with or without ID.

The ID is a smaller box, with two opposite walls designed as frames capable of allocating miniplates. Each position can contain two mini-plates in the vertical direction, while there are up to 6 horizontal positions, i. e. up to 12 mini-plates may be loaded inside the ID. Figure 2 shows a horizontal cross section of the central irradiation channel at half height, where both the EB and the ID may be seen. It is 30cm high, less than the total active length, being the mini-plates still shorter (see Figure 3).

A block model was chosen to represent the assembly EB + ID, which resulted from the compromise between a reasonable degree of spatial detail for neutron flux and economy in mesh points for a 3-D flexible and fast model of the whole reactor.

Figure 4 shows a diagram of the central irradiation channel with its seven blocks, but only four different materials. Each block is essentially a mixture of H₂O and Al except block IV which contains, besides, the meat material, constituted at present by an alloy of HEU (90%)U-Al, to be soon replaced by a LEU target. XS for each block were also generated with WIMS. The wide variety of XS enables calculations for the following different situations:

- **EB** OUT, **ID** OUT
- EB IN, ID OUT
- EB IN, ID-6 mini-plates IN
- EB IN, ID-12 mini-plates IN

3 RESULTS

3.1 Critical states

Table 2 summarises some results for C93_EOC and C94_BOC. All of them correspond to critical states with CR inserted as indicated, so that the experimental reactivity value is $\rho_{exp} = 0$.

It should be taken into account that the CR insertion value reported by the facility has a significant uncertainty, which could modify its calculated reactivity worth in up to 50pcm per each cm of error in the insertion position.

3.2 Reactivity worths associated to the irradiation device

According to the results contained in Table 2 and to non-critical state results, some reactivity worths were obtained as shown in Table 1. Experimental values are also shown in the same table, which have been extracted from /1/, and converted from \$ to pcm using $\beta_{eff} = 814$ pcm.

3.3 Core reactivity excess

Some discrepancies were observed between calculated and measures reactivity excess. They are mainly due to interaction among CR.

Figure 5 shows several calibration curves for one of the four CR (CR-4). Three out of four curves correspond to calculations being the fourth obtained experimentally. They all represent the (positive) reactivity worth inserted when a fraction of CR-4 is withdrawn.

According to /1/, the experimental calibration values (EXP) were obtained using CR-2 as compensating rod, i.e., CR-2 is gradually inserted as CR-4 is withdrawn so that the reactor remains (almost) critical. On the other hand, CR-4 calibration has been calculated for the following CR-2 situations:

- CR-2 fully inserted during the whole calibration procedure (CR-2-IN)
- CR-2 fully withdrawn during the whole calibration procedure (CR-2-OUT)
- CR-2 with increasing insertion in order to compensate withdrawal of CR-4 (CR-2-COMP).

The cases CR-2-COMP and EXP only differ in that the first one is the result of calculation and the second one is measured. CR-2 and CR-4 move equivalently for both cases, while the remaining CR are in fixed positions, namely CR-1 51.4% inserted and CR-3 fully withdrawn.

Comparison between cases CR-2-OUT and CR-2-IN shows that CR-4 reactivity worth may be significantly different according to CR-2 insertion, and the value corresponding to CR-2-OUT (some 1200 pcm smaller than that corresponding to the CR-2-IN case) should be used for reactivity excess calculations. CR-4 reactivity worth obtained in /1/ is 4046 pcm, while the calculated value is 3884pcm, with a calculation-measurement discrepancy of 162pcm (around 4% smaller for the calculated value).

The calculated reactivity excess for C94_EOC (cold, without Xe and long shutdown Sm concentration) was $\rho_{CALC} = 5115 pcm$, while the corresponding indirect measured value is $\rho_{EXP} = 5745 pcm$. Their discrepancy is due to both a calculation-measurement discrepancy and the interaction effect between measured and compensating CRs, being a reasonable value for C94_BOC reactivity excess $\rho = 5345 pcm$.

The irregular variations observed in CR-2-COMP curve are due to the fact that the partition in 10 axial regions does not always enable an exact representation of the fraction of CR inserted. When this is the case, CITVAP uses a mixture of XS, thus producing such artificial irregularities.

3.4 Effect of the presence of mini-plates on the neutron flux.

Taking C94-BOC (cold with CR fully withdrawn) as a reference state, the effect on the neutron flux due to the presence of 12 mini-plates (some 12g ²³⁵U) in the central irradiation channel was analysed. Figure 6 shows the space dependence of neutron thermal flux inside the ID in arbitrary units, showing its highest value at the cell midpoint and significantly decreasing values at each of the mini-plate locations (note cell symmetry), with similar absolute value.

Figures 7 and 8 correspond to reactor calculations, with a homogenised block IV (so that no flux fine structure can be observed inside the ID). Figure 7 shows thermal flux along a perpendicular to the mini-plates crossing the highest flux point at half height. The flux peak is relatively smaller with the ID inserted. Figure 8 shows flux depression in the fuel region.

3.5 Evaluation of the maximum specific power in the mini-plates.

The case of an ID loaded with 12 mini-plates is here considered. The following values were obtained:

Internal power peaking factor in block IV =1.0877

Total power in block IV = $3.8135 \times 10^4 \text{ W}$

Average power per mini-plate = $3.178 \times 10^3 \text{ W/ mini-plate}$

Average heat flux = 45.27 W/cm^2

Maximum heat flux = 49.25 W/cm^2

4 CONCLUSIONS

4.1 Validation of the calculation model

The model proposed has proved to be adequate for the evaluation of core criticality for different configurations, being the uncertainty smaller than 170 pcm, and probably less than that if CR insertion values are properly corrected.

On the other hand, both EB and ID reactivity worths have been calculated. The results obtained depend on the method used, be it either the change of XS or of CR position. The discrepancy between both methods lies within a range of 70 to 85pcm; this could be due to modelling limitations (in the representation of small variations in CR position) as well as to already mentioned uncertainties in the reading of insertion values and even to the β_{eff} value used. Anyway, calculated values agree with measured values within the uncertainty range. The validation carried out proves that the model is also adequate for configurations having the ID inserted.

4.2 Compliance with Argentine Nuclear Regulatory Standards

According to Argentine Nuclear Regulatory Standards, each time a new experiment or a design modification is foreseen in a research reactor, the ARN intervention is required. The ARN evaluates safety related effects of such modifications in order to issue the corresponding authorisation, taking into consideration its own information as well as that submitted by the installation itself.

In this report compliance with operational limits and conditions is verified as well as conditions related to fixed experiments contained in Standard AR 4.2.2 /7/.

The following items were verified:

a) Reactivity margins

Under the most reactive condition:

- The safety factor associated to safety rods $F = \frac{\Delta \rho_{safety rods}}{\Delta \rho_{react.\,excess}}$ shall be greater than 1.5.
- The reactor shall be maintained subcritical with a shutdown margin greater than 3000 pcm (4 safety rods fully inserted).
- The reactor shall be maintained subcritical with a shutdown margin greater than 1000 pcm even with the most reactive safety rod withdrawn ("stuck rod criterion").
- For any fixed experiment its reactivity worth shall be smaller than 1200 pcm.

b) Cooling conditions

The reactor cooling system shall be designed so as to provide adequate core cooling under any operational condition.

Reactivity margins have been here verified independently, having the installation provided its own results as required by the ARN.

As regards target cooling, it has been verified that:

- calculated heat flux at the mini-plates is smaller than its design value,
- CNEA measured values in a thermal-hydraulic test at a low pressure loop guarantee the necessary flow for mini-plate cooling.

It was also verified that an undesired reactivity insertion, due to a mis-management of the ID, is covered by the reactivity accident analysed in the SAR.

Finally, it was evaluated that the ID materials will not generate, under normal operation conditions, any chemical reaction, which could lead to explosions or increase corrosion.

4.3 Models proposed for LEU targets.

CNEA is presently considering some options to replace HEU by LEU targets. Such new targets are geometrically equivalent but they have a LEU UAl_2 -Al meat / 6 /. Test mini-plates have been already fabricated having the same dimensions as the present mini-plates, with a mass density of 3 g/cm^3 , which results in 1.4 g^{235} U/mini-plate. With such target type, an ID having the same design as the one here analysed can also be used without any difficulty.

A second option consists of a new device having 4 targets each of them constituted by a (LEU) U-metal foil. These 4 targets are arranged in two pairs of vertically aligned cylinders. Figure 9 shows a horizontal cross section of the new device at its active region. Figure 10 shows a possible modelling in three blocks of two different homogenised materials. Using adequate XS the results obtained are consistent with those for HEU targets. No experimental values are available yet.

5 REFERENCES

- /1/ Measurement of neutron parameters for the RA-3 reactor during start up (May 2000). A. Gómez, G. Estryk, D. Roqueta. CNEA.C.RCN.ITE.136.
- /2/ A summary of WIMSD4 input options, M.J. Halsall, AEEW M 1327, 1980.
- /3/ Nuclear reactor core analysis code CITATION, T.B. Fowler, D.R. Vondy, G.W. Cunningham, ORNL-TM-2496, 1971.
- /4/ CITVAP 3.1. MTR PC 2.6 User's Manual. July 1995.
- /5/ MTR_PC 2.6 User's Manual. July 1995.
- **/6/** Development of LEU targets of UAl2 for 99Mo production. C.A. Kohut, G.C. Rossi, E.E. Canil, D.G. Podestá, F.D. Canil, J. Falcone, M.A. Restelli, M. de la Fuente, P. Adelfang. UACN-CAC. CNEA, 1999.

6 TABLES

 Table 1: EB and ID Reactivity worth

Reactivity worth of:	Changing:	ρ ^{calc.} (pcm)	ρ exp. (pcm)
EB	XS	161	170 ± 80
EB	CR insertion	232	
ID (12g ²³⁵ U)	XS	783	863 ± 80
ID (12g ²³⁵ U)	CR insertion	867	

Table 2: Calculated reactivity for some critical states.

CORE	State / Poisons		CR fraction inserted	Central Irrad. Channel	ρ ^{calc.} (pcm)
N93 EOC	НОТ	C/Xe and equil Sm.	0 0 0 0.341	EB IN	-11
				ID-6m-pl IN	
N94 BOC	COLD	S/Xe and long shut down Sm	0.514 0 0 1	EB IN	100
				ID OUT	
N94 BOC	COLD	S/Xe and long shut down Sm	0.470 0 0 1	EB OUT	171
N94 BOC	COLD	S/Xe and long shut down Sm	0.712 0 0 1	EB IN	16
				ID-12m-pl IN	
N94 BOC	COLD	S/Xe and long shut down Sm	0.624 0 0 1	EB IN	71
				ID-6m-pl IN	
N94 BOC	COLD	S/Xe and long shut down Sm	0 0 0.524 1	EB IN	42
				ID OUT	

7 FIGURES

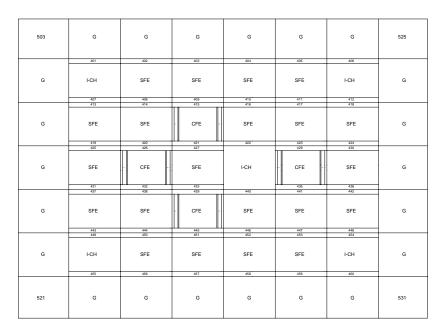


Figure 1: Core horizontal cross section at active half height.

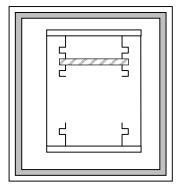
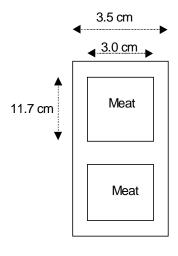


Figure 2: Horizontal cross section (active half height) of the central irradiation channel where EB and ID are located



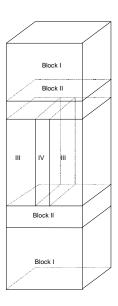
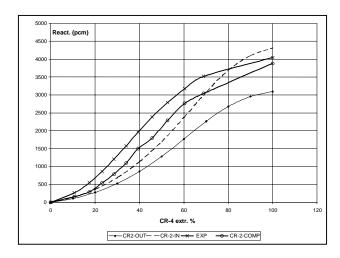


Figure 3: Arrangement of two mini-plates

Figure 4: Block model for the central irradiation channel (act. length 61.5cm)



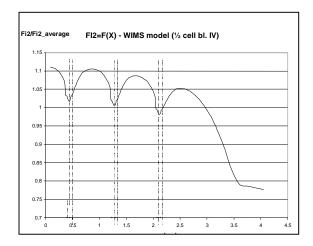
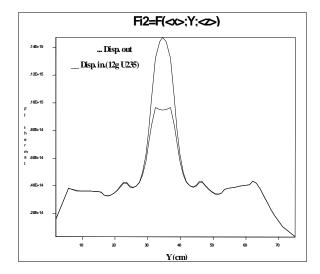


Figure 5: Calibration curves for CR-4.

Figure 6: Relative neutron thermal flux inside the ID (12 mini-plates. "Meat" regions for each mini-plate are indicated.



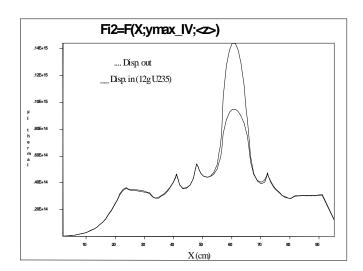


Figure 7: Neutron thermal flux along a perpendicular to the mini-plates with and without ID.

Figure 8: Neutron thermal flux along a parallel to the mini plates with and without ID

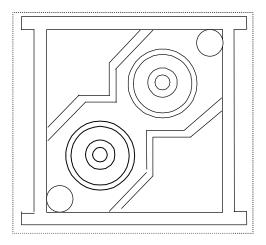


Figure 10: Cross section of the new ID (perpendicular to its main axis).

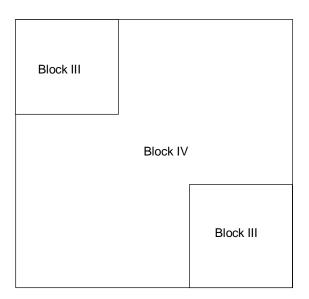


Figure 11: Cross section of the block model used for the central irradiation channel.